

This Page Is Inserted by IFW Operations  
and is not a part of the Official Record

## **BEST AVAILABLE IMAGES**

Defective images within this document are accurate representations of the original documents submitted by the applicant.

Defects in the images may include (but are not limited to):

- BLACK BORDERS
- TEXT CUT OFF AT TOP, BOTTOM OR SIDES
- FADED TEXT
- ILLEGIBLE TEXT
- SKEWED/SLANTED IMAGES
- COLORED PHOTOS
- BLACK OR VERY BLACK AND WHITE DARK PHOTOS
- GRAY SCALE DOCUMENTS

**IMAGES ARE BEST AVAILABLE COPY.**

**As rescanning documents *will not* correct images,  
please do not report the images to the  
Image Problem Mailbox.**



**Technical Language Service**

Translations From And Into Any Language

**JAPANESE / ENGLISH TRANSLATION OF**

**Japanese Patent Application JP 9 – 239883 A**

**Microbubble-Containing Laminated Polyester Film and  
Color-Printer Image-Receiving Paper Comprising Said Film**

**Your Ref: 050703-07**

**For: Eastman Chemical Company**

---

9109 Eagle Hills Dr . Las Vegas NV 89134 . ph: 866-722-7990 (toll-free) . fax: 702-233-5835 . email: sales@tls-translations.com

(19) Japanese Patent Office (JP)

(11) Unexamined Patent Application No:

**(12) Unexamined Patent Gazette (A)**

**Kokai 9-239883**

(43) Date of Publication: September 16, 1997

(51) Int. Cl. <sup>7</sup>	Class. Symbols	Internal Office Registration Nos.:	FI	Technical Classification Field
B 32 B 5/18			B 32 B 5/18	
B 29 C 55/12			B 29 C 55/12	
B 32 B 27/00			B 32 B 27/00	F
// B 29 K 67:00 105:04				
B 29 L 9:00				

Request for Examination: Not yet submitted

Number of Claims: 3 FD

(Total of 9 pages [in the original])

(21) Application No.: 8-83197

(22) Date of Filing: 12 March 1996

(71) Applicant: Diafoil Hoechst Co., Ltd.  
3-2-4 Shiba, Minato-ku,  
Tokyo

(72) Inventor: Takashi Hibiya  
c/o Central Research Center,  
Diafoil Hoechst Co., Ltd.,  
347 Inokuchi, Sakata-gun,  
Shiga Prefecture

(74) Agent: Kazuhiko Okada,  
Patent Attorney

**(54) [Title of the Invention]**

**Microbubble-containing Laminated Polyester  
Film and Color-Printer Image-receiving Paper  
Comprising Said Film**

**(57) [Summary]**

**[Object]** To provide an industrially advantageous microbubble-containing laminated polyester film that can be fabricated with high productivity and allows the cost of the raw material polyester to be reduced by employing regenerated polyester and using a relatively small amount of fluorescent whitening agent to achieve a sufficient degree of surface whitening, thereby reducing the amount of fluorescent whitening agent used.

**[Means of Achievement]** The present invention comprises a film in which a polyester layer A, which is an outermost layer, is laminated on at least one side of a microbubble-containing polyester layer B; the polyester layer B has a density of 0.40 to 1.30 g/cm<sup>3</sup> and

contains 5 to 45 wt% of a thermoplastic resin which is incompatible with the polyester; the polyester layer A contains 0.3 to 30 wt% white pigment and 0.01 wt% or more of a fluorescent whitening agent in a higher concentration than in the layer B; and the surface of the polyester layer A has a *b* value of +3.0 or less and a covering power of 0.3 or more.

#### **[Claims]**

**[Claim 1]** A microbubble-containing laminated polyester film, which is characterized in that a polyester layer A, which is an outermost layer, is laminated on at least one side of a microbubble-containing polyester layer B; the polyester layer B has a density of 0.40 to 1.30 g/cm<sup>3</sup> and contains 5 to 45 wt% of a thermoplastic resin which is incompatible with the polyester; the polyester layer A contains 0.3 to 30 wt% white pigment and 0.01 wt% or more of a fluorescent whitening agent in a higher concentration than in the layer B; and the surface of the polyester layer A has a *b* value of +3.0 or less and a covering power of 0.3 or more.

**[Claim 2]** The microbubble-containing laminated polyester film according to Claim 1, wherein the polyester layer B contains regenerated polyester.

**[Claim 3]** An image-receiving paper for color printers, which is characterized in that the paper comprises the microbubble-containing laminated polyester film as described in Claim 1 or 2.

#### **[Detailed Description of the Invention]**

##### **[0001]**

**[Technological Field of the Invention]** The present invention relates to a microbubble-containing laminated polyester film, and color printer image-receiving paper comprising said film.

##### **[0002]**

**[Prior Art]** Biaxially oriented polyester films are used across a wide range of application as an industrial material since they are well-balanced across their various physical properties and

are exceptional in terms of their cost performance. Microbubble-containing polyester films, formed into films using polyesters compounded with an incompatible thermoplastic resin, have been employed as white opaque films which are light in weight and are furnished with a cushioning property; examples of such applications include color printer image-receiving paper or other synthetic paper, magnetic cards, and the like. It is especially when these films are used as image-receiving paper that a fluorescent whitening agent and white pigment are compounded in the films in order to increase their whiteness so as to improve the vividness of the image, among other reasons.

**[0003]** However, effective use must be made of the fluorescent whitening agents and white pigments in order to keep the costs of the white opaque films to a minimum. Particularly when regenerated polyester is used as the polyester raw material in order to lower costs by a substantial margin, the need to avoid a decrease in the degree of whiteness caused by using regenerating polyester requires the fluorescent whitening agent and white pigment to be used in large amounts, which strongly dictates that these additives be used particularly effectively.

**[0004]**

**[Problems Which the Invention Is Intended to Solve]** With the foregoing problems of prior art in view, it is an object of the present invention to provide an industrially advantageous microbubble-containing laminated polyester film that can be fabricated with high productivity and allows the cost of the raw material polyester to be reduced by employing regenerated polyester and using a relatively small amount of fluorescent whitening agent to achieve a sufficient degree of surface whitening, thereby reducing the amount of fluorescent whitening agent used; and to provide a color printer image-receiving paper which comprises this film.

**[0005]**

**[Means Used to Solve the Above-Mentioned Problems]** As a result of conducting various investigations into means for achieving the aforementioned objective, the present inventors perfected the present invention by making expert use of a laminate structure to gain the insight necessary to enable the aforementioned objective to be readily achievable.

**[0006]** The present invention was perfected as a result of further investigations being performed based on the aforementioned insight, and comprises a first aspect, which consists in a

microbubble-containing laminated polyester film characterized in that a polyester layer A, which is an outermost layer, is laminated on at least one side of a microbubble-containing polyester layer B, the polyester layer B has a density of 0.40 to 1.30 g/cm<sup>3</sup> and contains 5 to 45 wt% of a thermoplastic resin, which is incompatible with the polyester, the polyester layer A contains 0.3 to 30 wt% white pigment and 0.01 wt% or more of a fluorescent whitening agent, which is a higher concentration than in the polyester layer B, the *b* value on the surface of the polyester layer A is +3.0 or less, and the covering power is 0.3 or more; and a second aspect, which consists in a color printer image-receiving paper characterized in that the paper is formed from the aforementioned minute air-bubble containing laminated polyester film.

[0007]

**[Embodiments of the Invention]** The present invention shall be described in detail hereunder. The microbubble-containing laminated polyester film of the present invention (abbreviated as "laminated film" hereunder) is formed by laminating a polyester layer A (abbreviated as "layer A" hereunder), which comprises an outermost layer, on at least one side of a microbubble-containing polyester layer B (abbreviated as "layer B" hereunder).

[0008] The polyester which constitutes the laminated film of the present invention is obtained with the principal starting raw materials being an aromatic carboxylic acid, or an ester of same, and a glycol, with 80% or more of its repeating structural units being ethylene terephthalate units or ethylene-2,6-naphthalate units. A third component may be included provided that the conditions do not deviate from the aforementioned range.

[0009] Other than terephthalic acid and 2,6-naphthalenecarboxylic acid, one or two or more of the following examples may be used for the aromatic carboxylic acid component: isophthalic acid, phthalic acid, adipic acid, sebacic acid, oxycarboxylic acid (e.g. *p*-oxyethoxybenzoic acid) and the like. Other than ethylene glycol, one or two or more of the following examples may be used for the glycol component: diethylene glycol, propylene glycol, butanediol, 1,4-cyclohexanedimethanol, neopentyl glycol and the like.

[0010] In the present invention, it is preferable to use various regenerated polyesters as the raw material polyester from the perspectives of raw material cost as well as environmental protection, which prescribes a reduction in the amount of scrap that needs to be treated after being discarded following the film manufacturing process. Examples of regenerated polyesters include those

recovered from magnetic cards, PET bottles and the like, as well as those obtained by a direct polymerization process. It is of course possible to use regenerated raw materials of microbubble-containing polyester films as such (e.g. spent products, trimmed film edges discarded after the film manufacturing process, and the like). These films, which are described hereunder, are obtained by making a film from polyester compounded with an incompatible thermoplastic resin.

[0011] When regenerated polyester is used in layer B in the laminated film of the present invention, the fact that layer B contains air bubbles and is covered by layer A means that it does not make any difference whether large particles are contained in the raw material or whether the coloration is in a correctable range. Accordingly, there is considerable latitude in the allowable range of regenerated polyester that can be used in the layer B, which offers significant cost-related benefits as well. Since the regenerated raw material polyester in the aforementioned microbubble-containing polyester film itself contains a thermoplastic resin which is incompatible with polyester, it is preferable for it to be compounded only with the raw material of the layer B.

[0012] Although the aforementioned regenerated polyester is commonly obtained in the form of regenerated chips after pulverization and melt extrusion, it does not necessarily have to be obtained in chip form; the polyester can be pulverized and then charged directly into a vented biaxial extruder together with chips of new polyester. The amount of the aforementioned regenerated polyester used will depend on the degree of coloration as well as other factors, with an amount of no greater than 60% normally being preferable.

[0013] The lower limit of the intrinsic viscosity of the polyester which constitutes the layer B during the film step is normally 0.45 or higher, and preferably 0.50 and higher. If the film has an intrinsic viscosity of less than 0.45, it will tend to rupture during the manufacturing step and the size of the air bubbles will tend to become irregular, thereby hindering control over density and leading to a decline in productivity. On the other hand, the upper limit should normally be 1.0 or less, insofar as productivity during the manufacturing of the polyester is concerned.

[0014] The intrinsic viscosity of the polyester which constitutes the layer A during the film stage may be the same as, or different from, that for the layer B; however, it is preferable for the layer A, which is a surface layer, to have a higher intrinsic viscosity than the layer B, which is an inner layer, due to the fact that the film will be prevented from rupturing during manufacture and particles will be less likely to fall off the film surface.



[0015] The layer B is obtained by making a film from a polyester compounded with an incompatible thermoplastic resin. Specific examples of the thermoplastic resin incompatible with the polyester include polyethylene, polypropylene, polymethylpentene, polymethylbutene and other polyolefins, as well as polystyrenes, polycarbonates, polyphenylene sulfides, liquid crystal polyesters and the like. Among the above examples, polypropylene is preferable from the perspectives of cost and productivity.

[0016] It is preferable for the aforementioned polypropylene to be a crystalline polypropylene homopolymer in which the propylene unit content is normally 95 mol% or greater, and preferably 98 mol% or greater. When a noncrystalline polypropylene is used, the polypropylene will bleed out on the surface of the amorphous polyester sheet during the film manufacturing process, and when the film comprises a two-layered structure from the layers A and B, the layer B will tend to soil the surfaces of the cooling drums and stretching rolls. Furthermore, if the polypropylene is a copolymer having more than, e.g., 5 mol% ethylene units in addition to the propylene units, the state of the individual air bubble formation will tend to be unsatisfactory.

[0017] The melt flow index (MFI) of the aforementioned polypropylene is normally selected from a range of 0.5 to 30 g/10 min, and preferably from 1.0 to 15 g/10 min. When the MFI is less than 0.5 g/10 min, the air bubbles which are formed will become too large, and rupturing will tend to occur more often during stretching; and when the MFI is greater than 30 g/10 min, the clips on the tenter, which is the stretching apparatus, will tend to come loose, and the density will become less uniform over time, thereby hindering density control, and adversely affecting productivity as a result. In the following description, polypropylene shall be used to represent a thermoplastic resin which is incompatible with polyester.

[0018] The amount of polypropylene contained in the polyester must be 5 to 45 wt%, preferably 5 to 35 wt% and even more preferably 10 to 25 wt% as a ratio relative to the total amount including the polyester. If the polypropylene content is less than 5 wt%, too few air bubbles will form in the film, making it impossible to manufacture a laminated film which is sufficiently light in weight and which has a good cushioning property. On the other hand, if the polypropylene content exceeds 45 wt%, the mechanical strength and thermostability of the laminated film will be inadequate, and the surface of the laminated film will become too rough. Such a laminated film would be unsuitable as an image-receiving paper, in which texture, vividness and other

printing properties are required. Furthermore, a polypropylene content exceeding 40 wt% will give rise to productivity-related drawbacks, such as frequent film rupturing during stretching.

[0019] In the present invention, it is preferable for the polyester which constitutes the layer B to contain a surfactant, because the density and cushioning property of the layer B can then be kept to within a desired range by controlling the size of the air bubbles which are formed during the film forming (stretching) process.

[0020] Examples of the aforementioned surfactant include anionic, cationic, amphoteric, and non-ionic variants, with the non-ionic surfactants, and silicone-based ones in particular, being preferable among these. Specific examples of silicone-based surfactants include organo-polysiloxane-polyoxyalkylene copolymer, an alkenylsiloxane, which contains a polyoxyalkylene side chain and the like. The surfactant content should normally be 0.001 to 1.0 wt% and preferably 0.01 to 0.5 wt%. If the surfactant content exceeds 1.0 wt%, the surfactant will no longer have any effect, and other difficulties will be presented, such as problems with the extruder, degradation in the polyester and the like.

[0021] The layer B must have a density, which is 0.40 to 1.30 g/cm<sup>3</sup>, preferably 0.60 to 1.20 g/cm<sup>3</sup>, and even more preferably 0.70 to 1.10 g/cm<sup>3</sup>. If the density exceeds 1.30 g/cm<sup>3</sup>, then the layer B will contain too few air bubbles and will tend to lose its cushioning property, which is a characteristic of the laminated film of the present invention. On the other hand, if the density of the layer B is less than 0.40 g/cm<sup>3</sup>, then the mechanical strength and thermostability of the laminated film will be inadequate, and other adverse effects will be presented relating to its production continuity and the quality of the film as an image-receiving paper.

[0022] It does not matter whether the microbubbles are present in the layer A in large quantities or are virtually absent, but their density should normally be 0.40 g/cm<sup>3</sup> or more, and preferably 0.60 to 1.50 g/cm<sup>3</sup> or more. If the density of the layer A is less than 0.40 g/cm<sup>3</sup>, then the surface of the layer A will become exceedingly rough, and when the laminated film is used as an image-receiving paper, the texture of the printed image will be lost and quality will also decrease.

[0023] The most significant characteristic of the laminated film of the present invention is that the layer A, which contains a fluorescent whitening agent in a high concentration as well as a white pigment in a specific concentration, is laminated on either or both sides of the layer B, which either contains virtually no fluorescent whitening agent or just a relatively small amount, in order for its properties as an image-receiving (printing) paper in particular to be enhanced.

[0024] Specifically, in the microbubble-containing laminated polyester film of the present invention, the layer A, which is an outermost layer, must contain 0.01 wt% or more of a fluorescent whitening agent, in a higher concentration than in the layer B, and 0.3 to 30 wt% white pigment, and the *b* value in the surface of the layer A of the laminated film must be +3.0 or less, and the covering power must be 0.3 or more. A figure of greater than +3.0 for the aforementioned *b* value will affect the color tone of the printed character images. It is preferable for the *b* value to be +1.0 or less, and more preferably -1.0 or less. If the aforementioned covering power is less than 0.3, the light blocking effect will be inadequate, and the vividness of the printed character images will suffer. The quality of the film as an image-receiving paper will be impaired. It is preferable for the covering power to be 0.5 or higher.

[0025] As described in the foregoing, it is necessary for the layer A to have a fluorescent whitening agent content of 0.01 wt%, and for the concentration thereof to be higher than for the layer B. If the fluorescent whitening agent content is less than 0.01 wt%, then it will become difficult to attain the aforementioned *b* value. Accordingly, the layer A is made to contain a higher concentration of fluorescent whitening agent than the layer B. In other words, by using a novel fluorescent whitening agent only in the layer A, which is a surface layer, the effect of its use will typically increase. To put it differently, the amount of fluorescent whitening agent will be reduced over the entire laminated film.

[0026] The amount of fluorescent whitening agent contained in the layer A is preferably within the range of 0.04 to 0.30 wt%. It is preferable for the relationship between the fluorescent whitening agent in the layers A and B to be expressed as: Layer A concentration (wt%) > Layer B concentration (wt%) + 0.03 (wt%). Examples of suitable fluorescent whitening agents include "Ubitec", which is a brand name of Ciba-Geigy AG, and "OB-1", which is a brand name of Eastman Kodak Corporation.

[0027] As has been described in the foregoing, it is necessary for the white pigment content in layer A to be 0.3 to 30 wt%. If the white pigment content is less than 0.3 wt%, then the aforementioned degree of covering power cannot be achieved, while a content in excess of 30 wt% will cause the white pigment to aggregate and coarse protrusions to form, among other problems. When using two or more kinds of white pigment, the total content should be adjusted to come within the aforementioned range.

[0028] The content of white pigment in the layer A is preferably 0.5 to 20 wt%, and even more preferably 1.0 to 20 wt%. When using two or more types of pigment, the total pigment content should be adjusted to come within the aforementioned range. Titanium dioxide, barium sulphate and the like are suitable examples of white pigments, which can be used. When using two or more kinds of white pigment, it is preferable for at least one to be either titanium oxide or barium sulfate. The covering power and degree of whiteness of the laminated film may be enhanced by having the layer B contain the same or different white pigments as the layer A. It is preferable for the white pigment content of the layer B to be in similar range to the layer A.

[0029] The average particle diameter of the white pigment is normally 5.0  $\mu\text{m}$  or less and is preferably selected from a range of 0.01 to 3.0  $\mu\text{m}$ . If the average particle diameter exceeds 5.0  $\mu\text{m}$ , then the surface of the laminated film will become too rough, which will lead to various problems such as a reduction in the quality of the printed image and the white pigment tending to come off the surface of the laminated film.

[0030] In the present invention, the fluorescent whitening agent, white pigment and other additives may be added during the polyester synthesis reaction or directly to the polyester. When the additive is added during the polyester synthesis reaction, it is preferable to use a method whereby a slurry in which ethylene glycol or the like has been dispersed is added at any stage during polyester synthesis. When the additive is added directly to the polyester, it is preferable to use a method which involves using a biaxial kneader extruder to blend the additive with the polyester, with the additive being either in a dry state, or dispersed in water or an organic solvent that has a boiling point of 200°C or less, to form a slurry.

[0031] An effective method for controlling the additive content involves using the aforementioned method to adjust the master raw material, which contains the additive in a high concentration, and then controlling the additive content at the time of film manufacture by diluting the master raw material with a raw material that essentially contains no additives. The white pigment may be subjected to any desired pretreatment such as crushing, dispersing, classifying, filtration and the like. In the present invention, the polyester or polypropylene may be compounded with an antioxidant, thermostabilizer, lubricant, antistatic agent, dye, pigment or other additive as needed.

[0032] The laminated film of the present invention is normally manufactured by the following coextrusion method. First, the polyester raw materials, which constitute each layer are fed to a

coextrusion lamination extrusion apparatus. In other words, the polyester is laminated by using two or more extruders and a feedblock or multi-manifold having two or more layers, and is then extruded through a slit die as a molten sheet. For films which have an A/B/A layer structure, each polyester raw material which constitutes the layer A or layer B is extruded from one extruder each, with the layer A melt line being divided during that process.

[0033] A metering feeder such as a gear pump is arranged on the divided melt line to control the flow rates for the polyester raw materials of the layer A, which are laminated above and below the layer B, thereby laminating the layer B on both of its sides. The thickness of each layer A can be controlled by the flow rate. As a matter of course, the metering feeder can be provided on a melt line for the layer B polyester raw material to effectively control its thickness.

[0034] Next, the molten sheet extruded from the die is solidified by being rapidly cooled to a temperature below a glass transition temperature thereof on a rotary cooling drum, to form a substantially amorphous unoriented sheet. In such a case, in order to increase the flatness of the sheet, the adhesion between the sheet and the rotary cooling drum must be increased, and in the present invention, an electrostatic pinning method and/or a liquid-coating pinning method are preferably adopted to achieve such a state.

[0035] Electrostatic pinning generally refers to a method in which a wire electrode is disposed over an upper surface of the sheet so as to extend in a direction perpendicular to the feed direction of the sheet, and a D.C. voltage of approximately 5 to 10 kV is applied to the electrode to produce a static charge in the sheet, thereby enhancing the adhesion between the laminated sheet and the drum. Liquid-coating pinning refers to a method in which the adhesion between the sheet and the rotary cooling drum is improved by uniformly applying a liquid on a part (for example, only regions which come into contact with the opposite edges of the sheet) or the whole of a surface of the drum. In the present invention, both methods may be used in combination, if required.

[0036] The resulting sheet is subsequently stretched in at least one direction to form a film. The microbubbles in the layer B of the laminated film of the present invention are produced in the aforementioned stretching process. Accordingly, it is necessary for the stretching to be conducted under suitable conditions in order to form microbubbles in a well-controlled manner and to suitably satisfy such requirements as strength and dimensional stability in the film; for this reason, the following biaxial stretching method and heat-treating method are suitably adopted.

[0037] First, the unstretched sheet is stretched in one (longitudinal) direction at a drawing temperature of normally 70 to 150°C, and preferably 75 to 130°C and at a draw ratio of normally 3.0 to 7 times, and preferably 3.2 to 6 times. Such stretching may be conducted by using a roll-type or tenter-type stretching machine. Next, the sheet is stretched in a direction (transverse direction) perpendicular to the first-stage direction at a drawing temperature of normally 75 to 150°C, and preferably 80 to 140°C, and at a draw ratio of normally 3.2 to 7 times, and preferably 3.5 to 6 times, to obtain a biaxially oriented film. The latter stretching may also be conducted by using a tenter-type stretching machine.

[0038] The stretching in each direction can also be conducted in two or more stages; however, in such cases it is preferable for the final draw ratio to come within the aforementioned range.

Alternatively, the aforementioned unstretched sheet can be subjected to a simultaneous biaxial stretching, so as to yield an area magnification of 10 to 40 times. A heat treatment is performed from 1 second to 5 minutes at 150 to 250°C, under conditions of an elongation of not more than 30%, a limited shrinkage or a constant length. Another method involves the heat treatment being performed after the biaxially stretched film is re-stretched in the longitudinal direction at a temperature of 110 to 180°C and a draw ratio of 1.05 to 2.0 times. In such instances, various procedures such as heat setting before longitudinal re-stretching, longitudinal relaxation after longitudinal re-stretching, fine longitudinal stretching before or after longitudinal re-stretching, or the like can be appropriately used. Similarly, the film can be re-stretched in the transverse direction. Various surface treatments can also be performed as needed during the film manufacturing processes.

[0039] The total length of the laminated film of the present invention is normally established as 20 to 250  $\mu\text{m}$ , and preferably 20 to 75  $\mu\text{m}$ . The thickness of the layer B is normally established as being 20% or more, and preferably 50 to 99 wt% relative to the total thickness. When the thickness of the layer B is less than 20%, the resulting laminated film will be insufficient with regard to its light weight, good cushioning property or other requirements, nor will the effect of reducing the amount of fluorescent whitening agent used, which is a characteristic of the present invention, be adequately exhibited.

[0040] The characteristics of the laminated film of the present invention allow it to be suitably applied to labels, recording paper, posters, planographic printing plates, wrapping materials or the like, and particularly to image-receiving paper for color printers.

[0041] The image-receiving paper for color printers of the present invention comprises a laminated film such as described in the foregoing, and the layer A, which is an outermost layer, constitutes the printing surface. The image-receiving paper has an exceptional image definition, vividness and image-receiving property, as voids are not present in the printed images, and can be manufactured inexpensively.

[0042]

**[Working Examples]** The present invention shall be described in detail hereunder with reference to practical examples; however, the present invention shall not be limited to these examples as long as the essence of the present invention is preserved. The evaluation methods used in the comparative and working examples are as shall be described below. In the working examples and comparative examples, "part(s)" represent "weight part(s)" in all cases; the MIF [sic] of the crystalline polypropylene chips used is 10 g/10 min; the average particle diameter of the titanium oxide is 0.3  $\mu\text{m}$ ; "OB-1", produced by Eastern Kodak Corp., is used as a fluorescent whitening agent; and "SH-193", produced by Dow Corning Toray Silicone Co., Ltd., is used as a silicone-based surfactant.

[0043] (1) Thickness of respective layers: a cross section of the film was observed by a transmission electron microscope (TEM). In other words, a small piece of the film was wrapped and embedded in an epoxy resin in which a hardening agent and an accelerating agent had been compounded. The resulting film was cut into a piece approximately 200 nm thick using an ultra-microtome, to form a sample for observation. A photomicrograph of a cross-section of the sample was taken by a transmission electron microscope ("H-9000"; manufactured by Hitachi Ltd.) to measure the thicknesses of the respective layers of the sample. In the measurement, the transmission electron microscope was adjusted so that the accelerating voltage was 300 kV and the magnification was in the range of 10,000 to 100,000 times according to the thickness of an outermost layer of the sample. The thickness measurement was conducted at fifty different points, with the 10 thickest and 10 thinnest values being discarded, and the average of the remaining 30 values being taken as the thickness.

[0044] (2) Film density ( $\text{g}/\text{cm}^3$ ): a square sample 10 cm  $\times$  10 cm in size was cut from an indeterminate portion of the film; the sample was weighed and the thickness thereof was measured by a micrometer at nine indeterminate points thereon to obtain an average thickness

value, which was used together with the weight to determine the weight per unit volume. Five measurements ( $n$ ) were taken, and the average thereof was calculated to obtain the film density. The thicknesses of the respective layers were measured by a transmission electron microscope, after which a surface layer of the film (layer A) was abraded by a microtome and the density of the surface layer (layer A) was measured using a density gradient tube; based on the resulting surface layer density, the densities of the other layers (layer B) were calculated.

[0045] (3) Whiteness ( $b$  value): the color tones ( $L$ ,  $a$ ,  $b$ ) of the film were measured by a color analyzer ("TC-1800MKII"; manufactured by Tokyo Denshoku Co., Ltd.) according to JIS Z-8722, with the whiteness of the film being represented by the  $b$  value. A larger positive (+)  $b$  value indicates that the film tended to be more yellow. The whiteness of the film was evaluated by the average of the  $b$  values measured at three points thereon.

[0046] (4) Covering power: a visual light was used to measure the transmission concentration using a "TD-904" Macbeth densitometer. The covering power value was determined as the average of measured values taken at five points. A larger covering power value indicates that the film has a lower light transmittance.

[0047] (5) Average particle diameter of pigment and particle size distribution value: a centrifugal sedimentation-type particle size distribution measuring apparatus ("SA-CP3"; manufactured by Shimadzu Seisakusho Co., Ltd.) was used for the measurements, and particle diameters having a cumulative volume fraction of 50% in the equivalent spherical distribution were taken as the average particle diameter.

[0048] (6) Intrinsic viscosity  $[\eta]$  of polyester (dl/g): one gram of polyester from which polymer components incompatible with polyester and pigments were removed, was dissolved in 100 ml of a mixed solvent comprising phenol and tetrachloroethane at a weight ratio of 50:50; the intrinsic viscosity of the polyester solution was measured at 30°C.

[0049] (7) Melt flow index (MFI) (g/10 min): the melt flow index of the film was measured according to JIS K-6758-1981. A high MFI value indicates a lower melt viscosity of the polymer.

[0050] (8) Effects of reducing the amount of fluorescent whitening agent "OB-1" used: the concentrations of the fluorescent whitening agent "OB-1" added to the raw materials of the films in all of the layers were compared with each other, with those films requiring a higher concentration of the fluorescent whitening agent relative to a reference of 5 wt% being graded with an "×" mark, while those having a lower concentration were graded with a "○" mark



[0051] (10) Evaluation of image-receiving property: the film was cut into an A4-size sheet on which images were printed by thermal transfer recording, using a color printer ("CX-5000"; manufactured by Sharp Corp.), and the quality of the image on the resulting hard copy was visually observed. The results obtained by observing the definition, void [presence] and vividness of the images were classified as thus: an "o" mark indicated a high quality image, a delta indicated a slightly inferior image, which was nonetheless useable, and an "x" mark indicated an image which would be problematic in practical application.

#### [0052] Comparative Example 1

16.7 wt% crystalline polypropylene chips, 2.5 wt% titanium oxide and 0.3 wt% silicone-based surfactant were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.69, after which the mixture was uniformly blended to prepare a polyester raw material A. 2.5 wt% titanium oxide was compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.69, after which the mixture was uniformly blended to prepare a polyester raw material B.

[0053] A co-extruded unstretched sheet having three layers made of two different raw materials was obtained by charging the aforementioned respective raw materials into separate extruders and melting them at 290°C, then introducing the resulting molten raw materials into a common die, extruding them through a slit die so that layers of the raw material A were melt-laminated onto both outer surfaces of an inner layer made of the raw material B, and cooling the laminated film using a 40°C cooling drum. The unstretched sheet was stretched in a longitudinal direction at a drawing temperature of 85°C at a draw ratio of 3.4 times, after which the sheet was stretched in the transverse direction at 110°C at a draw ratio of 3.2 times and then subjected to a heat treatment at 230°C for 5 seconds to result in a final biaxially oriented laminated film having a thickness of 50  $\mu\text{m}$  and a density of 0.97  $\text{g}/\text{cm}^3$ . The resulting laminated film was evaluated, and the results are shown in Tables 1 to 3. The laminated film had a high *b* value, which indicated a diminished image-receiving property.

#### [0054] Working Example 1

The same procedure as followed in Comparative Example 1 was conducted for the preparation of the raw material B to prepare a polyester raw material D, with the exception that

the amount of titanium oxide compounded was changed to 7.0 wt%. Separately, 7.0 wt% titanium oxide and  $5 \times 10^{-2}$  wt% fluorescent whitening agent were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.69, after which the mixture was uniformly blended to prepare a polyester raw material C. Subsequently, two 3-layer biaxially oriented laminated films which had the structure C/D/C were prepared in the same manner as defined in Comparative Example 1, with the results obtained from evaluating these films being shown in Tables 1 to 3. The laminated films had exceptional image-receiving properties.

#### [0055] Comparative Example 2

Film scraps produced during the manufacture of the laminated film of Comparative Example 1 were fed into a biaxial extruder and melt-extruded to yield regenerated raw material chips X. The polyethylene terephthalate in the regenerated chips had an intrinsic viscosity of 0.61.

[0056] 40 wt% of the aforementioned regenerated raw material X, 13 wt% crystalline polypropylene chips and 0.15 wt% fluorescent whitening agent were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.74, and then 2.5 wt% titanium oxide and 0.3 wt% silicone-based surfactant were further compounded therewith, after which the resulting mixture was uniformly blended to prepare a polyester raw material F. Separately, 7 wt[%] titanium oxide and  $5.0 \times 10^{-2}$  wt% fluorescent whitening agent were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.69, after which the mixture was uniformly blended to prepare a polyester raw material E. Subsequently, two 3-layer biaxially oriented laminated films which had the structure E/F/E were prepared in the same manner as defined in Comparative Example 1, and the results of evaluating the films are shown in Tables 1 to 3. The laminated films were disadvantageous in terms of their production cost because of the large amount of fluorescent whitening agent required, despite having an exceptional image-receiving property.

[0057] 40 wt% of the aforementioned regenerated raw material X and 13 wt% crystalline polypropylene chips were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.74, and then 2.5 wt% titanium oxide and 0.3 wt% silicone-based surfactant were further compounded therewith, after which the mixture was uniformly blended to

prepare a polyester raw material H. Separately, 7 wt[%] titanium oxide and  $10 \times 10^{-2}$  wt% fluorescent whitening agent were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.69, after which the mixture was uniformly blended to prepare a polyester raw material G. Subsequently, two 3-layer biaxially oriented laminated films which had the structure G/H/G were prepared in the same manner as defined in Comparative Example 1, and the results of evaluating the films are shown in Tables 1 to 3. The laminated films had exceptional image-receiving properties.

#### [0058] Comparative Example 3

20 wt% of the aforementioned regenerated raw material X, 13 wt% crystalline polypropylene chips and 0.3 wt% silicone-based surfactant were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.71, after which the mixture was uniformly blended to prepare a polyester raw material J. Separately,  $1.0 \times 10^{-2}$  wt% fluorescent whitening agent was compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.69, after which the mixture was uniformly blended to prepare a polyester raw material I. Subsequently, two 3-layer biaxially oriented laminated films which had the structure I/J/I were prepared in the same manner as defined in Comparative Example 1, and the results of evaluating the films are shown in Tables 1 to 3. The laminated films had a diminished image-receiving property.

#### [0059] Comparative Example 4

The same polyester raw material D as prepared in Working Example 1 was used in this comparative example. Separately, 35 wt% titanium oxide and  $5 \times 10^{-2}$  wt% fluorescent whitening agent were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.69, after which the mixture was uniformly blended to prepare a polyester raw material K. Subsequently, two 3-layer biaxially oriented laminated films which had the structure K/D/K were prepared in the same manner as defined in Comparative Example 1, and the results of evaluating the films are shown in Tables 1 to 3. The laminated films had a diminished image-receiving property.

**[0060] Comparative Example 5**

The same polyester raw material C as prepared in Working Example 1 was used in this comparative example. Separately, 3 wt% crystalline polypropylene chips and 7.0 wt% titanium oxide were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.69, after which the mixture was uniformly blended to prepare a polyester raw material L. Subsequently, two 3-layer biaxially oriented laminated films which had the structure C/L/C were prepared in the same manner as defined in Comparative Example 1, and the results of evaluating the films are shown in Tables 1 to 3. The laminated films had a diminished image-receiving property.

**[0061] Comparative Example 6**

The same polyester raw material C as prepared in Working Example 1 was used in this comparative example. Separately, 50 wt% crystalline polypropylene chips and 7.0 wt% titanium oxide were compounded with polyethylene terephthalate chips which had an intrinsic viscosity of 0.69, after which the mixture was uniformly blended to prepare a polyester raw material M. Subsequently, two 3-layer biaxially oriented laminated films which had the structure C/M/C were prepared in the same manner as defined in Comparative Example 1, and the results of evaluating the films are shown in Tables 1 to 3. The laminated films had a diminished image-receiving property.

[0062]

[Table 1]

	Density (g/cm <sup>3</sup> )		Thickness by layer (μm)		Content in film (wt%)	
	Layer A	Layer B	Layer A	Layer B	Layer A: TiO <sub>2</sub>	Layer B: PP
Comparative Example 1	1.44	0.92	2.5	45	2.5	16.7
Working Example 1	1.44	0.90	12.5	25	7	16.7
Comparative Example 2	1.44	1.05	2.5	45	7	19
Working Example 2	1.44	1.05	2.5	45	7	19
Comparative Example 3	1.40	0.95	2.5	45	0	16
Comparative Example 4	1.52	0.89	2.5	45	35	16.7
Comparative Example 5	1.44	1.33	2.5	45	7	3
Comparative Example 6	1.44	0.58	2.5	45	7	50

[0063]

[Table 2]

	Amount of OB1 added ( $10^{-2}$ wt%)			Film properties	
	In layer A	In all layers	Effect of reducing the amount of OB1	<i>b</i> value in layer A surface	Covering power in layer A surface
Comparative Example 1	0	0	–	–0.8	0.7
Working Example 1	5	2.5	○	–2.8	0.8
Comparative Example 2	5	14	×	–3.0	0.7
Working Example 2	10	1.0	○	–3.2	0.7
Comparative Example 3	1	0.1	○	–2.0	0.2
Comparative Example 4	5	0.5	○	–2.5	1.0
Comparative Example 5	5	0.5	○	–3.1	0.6
Comparative Example 6	5	0.5	○	–3.4	0.5

[0064]

[Table 3]

	Image-receiving properties		
	Definition of image quality	Voids in image	Vividness of image quality
Comparative Example 1	○	○	×
Working Example 1	○	○	○
Comparative Example 2	○	○	○
Working Example 2	○	○	○
Comparative Example 3	○	○	×
Comparative Example 4	×	×	○
Comparative Example 5	○	×	○
Comparative Example 6	×	×	○

**[Merits of the Invention]** The microbubble-containing laminated polyester film of the present invention has a layer that contains minute individual air bubbles, which allows the weight per unit volume of the film to be reduced and which provides a cushioning property, while the fact that the film contains a relatively thin laminated layer that contains a fluorescent whitening agent in a high concentration allows it to exhibit sufficient whiteness on the surface of said layer, and in particular, enables a highly satisfactory image quality to be realized when used as a paper in various printers; moreover, the film is exceptional in terms of cost performance, due to its effect of reducing the amount of fluorescent whitening agent needed.